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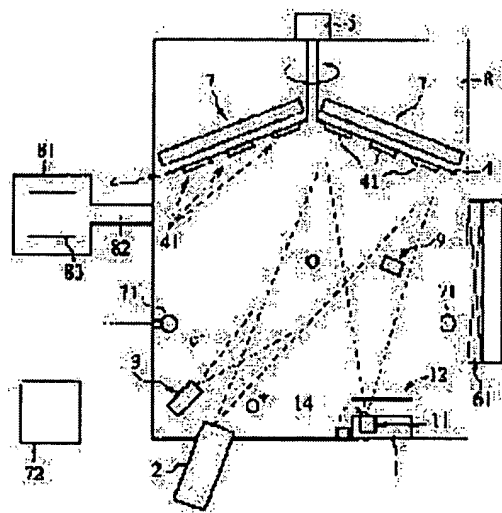
(72)Inventor : ESHITA TAKASHI
HIROSE TATSUYA
HIKOSAKA YASUMI

(54) METHOD FOR DEPOSITING DIELECTRIC THIN FILM

(57)Abstract:

PURPOSE: To deposit dielectric thin films having good quality by executing vapor deposition at a specific deposition speed in an active oxygen atmosphere at the time of irradiating a raw material evaporating source with electron beams to heat and evaporate the raw material and vapor-depositing the dielectric thin films on substrate surfaces.

CONSTITUTION: The inside of a vapor deposition chamber 8 is specified at about 1×10^{-5} to 1×10^{-3} Torr and the substrates 41 is heated to $\geq 200^\circ\text{C}$. The evaporating source 1 (SrTiO₃, etc.) in a crucible 1 is evaporated by irradiation with the electron beams, by which the dielectric thin films are formed on the substrates 41. At this time, oxygen is introduced into an ion gun 2 to form an oxygen atom current. This ion current is irradiated with the electron beams generated by an electron irradiation device 3, by which the ion current is electrically neutralized and an oxygen atom current is formed. The oxidized high-dielectric constant thin films or oxide ferroelectric thin films are deposited at a deposition rate of $\leq 0.2\text{nm/s}$ on the substrates 41 by irradiating the substrates with this oxygen atom current. As a result, the



adequate dielectric thin films are formed on the semiconductor substrates, etc.

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CLAIMS

[Claim(s)]

[Claim 1] The dielectric thin film deposition approach characterized by making an oxide quantity dielectric constant thin film or an oxide ferroelectric thin film vapor-deposit and deposit on a substrate as below the rate of sedimentation 0.2 [nm/s] under the ambient atmosphere containing active oxygen, such as an accelerated oxygen molecule, an oxygen radical, an oxygen atom, or oxygen plasma.

[Claim 2] The dielectric thin film deposition approach according to claim 1 characterized by carrying out the oxygen tension in an ambient atmosphere more than 10^{-4} [Torr].

[Claim 3] An oxide quantity dielectric constant thin film or an oxide ferroelectric thin film is SrTiO_3 , BaTiO_3 , TiO_2 , Ta_2O_5 , PbTiO_3 , and PbZrO_3 . Claim 1 characterized by being mixed crystal, such as either or this, or the dielectric thin film deposition approach given in two.

[Claim 4] Claim 1 characterized by for an electron neutralizing electrically the oxygen ion with which the accelerated oxygen molecule, the oxygen radical, the oxygen atom, etc. were accelerated by Ion Gun, and being generated, and irradiating more than 1×10^{13} [/cm² and s] to a substrate, 2, or the dielectric thin film deposition approach of three publications.

[Claim 5] Claim 1 characterized by generating the oxygen plasma at a vacuum evaporation room, 2, or the dielectric thin film deposition approach given in three.

[Claim 6] Claim 1 characterized by introducing into a vacuum evaporation room the active oxygen which is made to generate the oxygen plasma and is obtained at an oxygen plasma generating room, 2, or the dielectric thin film deposition approach given in three.

[Claim 7] Claim 1 which carries out substrate temperature more than 200 [°C], and is characterized by making an oxide quantity dielectric constant thin film or an oxide ferroelectric thin film vapor-deposit and deposit, 2, 3, 4, 5, or the dielectric thin film deposition approach of six publications.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Industrial Application] This invention relates to the approach of depositing a suitable dielectric thin film producing a small capacitor for example, to a semi-conductor substrate etc.

[0002] It will be necessary to make small the capacitor of a comparatively big capacity connected out of the chip, and it will need to be incorporated as an internal component of MMIC in the microwave integrated circuit (monolithic microwave integrated circuit:MMIC) for recent years, for example, cellular phones, etc. for a miniaturization or low-pricing.

[0003] Moreover, like dynamic random access memory (dynamic random access memory:DRAM), detailed-ization is needed with high integration, and it is necessary to produce the capacitor for storage maintenance in a small area.

[0004] Since it is such, desire of the implementation of how a dielectric constant can mass-produce a high thin film or a ferroelectric thin film is carried out.

[0005]

[Description of the Prior Art] Usually, an ingredient with the high dielectric constant for capacitors or a ferroelectric 3, for example, SrTiO₃, BaTiO₃, TiO₂, Ta₂O₅, PbTiO₃, and PbZrO₃ Or mixed crystal, such as this, etc. For example, the sputtering method (see "JP,58-194206,A", if it requires), a chemical-vapor-deposition method (if it requires) H. "Yamaguchi et al., Jpn.J.Appl.Phys.vol.32 No.9B, pp.4069-4073 (1993)" -- reference and a sol-gel method (if it requires) Therefore, it is produced by applying [reference] "JP,56-28408,A."

[0006] By the sputtering method in said Prior art, the dielectric thin film is formed by making it for example, low-pressure Ar ambient atmosphere, generating the plasma and generally, carrying out sputtering of the target within a vacuum housing, after setting the semi-conductor substrate which should counter the target which consists of a dielectric raw material, and should form a dielectric thin film.

[0007] In this case, there is comparatively little number of sheets of the semi-conductor substrate which can be processed at once since there is a limitation in the magnitude of a target, and it has a possibility of deteriorating the property of the device which a semi-conductor substrate front face is put to the ion of high energy etc., and is easy to damage, therefore is made in order to form a thin film in the plasma.

[0008] moreover, chemical vapor deposition (chemical vapor deposition:CVD) -- although material gas is made to react at an elevated temperature and a dielectric thin film is made to deposit on a semi-conductor substrate in law -- SrTiO₃ and BaTiO₃ etc. -- forming thin films, such as this, on mass-production level, since there are not Sr raw material with high vapor pressure and a Ba raw material in order to make it grow up -- in practice -- coming out -- it is difficult.

[0009] Furthermore, apply the emulsion or suspension which consists of an organic solvent and dielectric-materials impalpable powder on a semi-conductor substrate, and therefore evaporate an organic solvent to process this at the elevated temperature more than about 600 [**], and he makes a particle sinter, and is trying to obtain a desired dielectric thin film with a sol-gel method again.

[0010] Therefore, in the phase after forming a transistor, although the processing in an elevated temperature is indispensable, when an elevated-temperature process cannot be applied, for example, since a transistor deteriorates by applying an elevated temperature, this means cannot be adopted.

[0011] By the way, a lot of [faults / there are comparatively few faults looked at by the above mentioned all directions method, namely,] semi-conductor substrates can be made to deposit an oxide thin film at low temperature, and vacuum deposition is known as an approach with doing [little] damage to a semi-conductor substrate front face like the sputtering method.

[0012] However, the oxygen in the oxide thin film made to deposit when making an oxide thin film deposit, in order to carry out in a vacuum tends to escape from vacuum deposition, and a good thin film is not obtained.

[0013] The approach of vapor-depositing like for example, ion beam assistant vacuum deposition, there, while irradiating an oxygen beam at a thin film is also proposed.

[0014] However, it is [that the thin film for optics (see "JP,61-288064,A" if it requires), and the interlayer insulation film (see "JP,3-196526,A" if it requires) in an integrated circuit are generally only known for the thin film for capacitors as an oxide thin film formed with the application of ion beam assistant vacuum deposition since it is required that electrical characteristics' being good, especially specific inductive capacity should be high, and leakage current should be low, and], and there is no example about the thin film for capacitors.

[0015] Therefore, this invention persons are actually SrTiO₃, BaTiO₃, TiO₂, Ta₂O₅, PbTiO₃, and PbZrO₃ to the vacuum deposition indicated by said each patent official report. Although the capacitor was made as an experiment using the thin film, no thin film was able to fulfill high specific inductive capacity and the conditions of little leakage current.

[0016]

[Problem(s) to be Solved by the Invention] It enables it to mass-produce a thin film with the high dielectric constant which can be used for the capacitor for integrated circuits at comparatively low substrate temperature by this invention. For this reason, vacuum deposition is applied, electrical characteristics were excellent, namely, specific inductive capacity makes high deposition of a high dielectric thin film with little leakage current possible.

[0017]

[Means for Solving the Problem] This invention is selecting the rate of sedimentation and the oxygen tension at the time of deposition to a suitable value, when making a dielectric thin film deposit with the application of vacuum deposition with the compensation means of oxygen like ion beam assistant vacuum deposition, and it has been to the base to realize a dielectric thin film with the outstanding property which is equal to use as a configuration member of the capacitor for integrated circuits.

[0018] In the dielectric thin film deposition approach which depends on this invention from the above mentioned place (1) The accelerated oxygen molecule, an oxygen radical, an oxygen atom, Or [whether it is characterized by making an oxide quantity dielectric constant thin film or an oxide ferroelectric thin film (for example, SrTiO₃ thin film etc.) vapor-deposit and deposit on a substrate (for example, semi-conductor substrate 41) as below the rate of sedimentation 0.2 [nm/s] under the ambient atmosphere containing active oxygen (oxygen style), such as oxygen plasma, and] Or [0019] (2) or [that it is characterized by carrying out the oxygen tension in an ambient atmosphere more than 10⁻⁴ [Torr] in the above (1)] -- [0020] [or] (3) the above (1) or (2) -- setting -- an oxide quantity dielectric constant thin film or an oxide ferroelectric thin film -- SrTiO₃, BaTiO₃, TiO₂, Ta₂O₅, PbTiO₃, and PbZrO₃ any -- or [or / that it is characterized by being mixed crystal, such as this,] -- [0021] [or] (4) The oxygen molecule accelerated in the above (1), (2), or (3), An oxygen radical, or [that it is characterized by for an electron (for example, electron from electronic irradiation equipment 3) neutralizing electrically the oxygen ion with which the oxygen atom etc. was accelerated by Ion Gang (for example, Ion Gang 2) and being generated, and irradiating more than 1x10¹³ [/cm² and s] to a substrate] -- or [0022] (5) or [that it is characterized by generating the oxygen plasma at a vacuum evaporation room in the above (1), (2), or (3)] -- [0023] [or] (6) or [that it is characterized by introducing into a vacuum evaporation room the active oxygen which is made to generate the oxygen plasma and is obtained in the above (1), (2), or

(3) at an oxygen plasma generating room] -- [0024] [or] (7) In the above (1), (2), (3), (4), (5), or (6), substrate temperature is carried out more than 200 [**], and it is characterized by making an oxide quantity dielectric constant thin film or an oxide ferroelectric thin film vapor-deposit and deposit. [0025]

[Function] A diameter is able to be able to use a thing large-sized as deposition equipment for it, and to carry out batch processing of many semi-conductor substrates of 75 [mm] thru/or 100 [mm] to it therefore, since it is the same as carrying out the usual vacuum evaporatio for taking said means fundamentally.

[0026] Moreover, as for a different place from the usual vacuum evaporatio, since the amount of oxygen therefore contained in an oxygen exposure at a dielectric thin film is controllable, electrical characteristics can make a good dielectric thin film able to deposit, and can raise the crystallinity of a dielectric thin film by therefore making a substrate a heater at suitable temperature in the case of vacuum evaporatio.

[0027] Moreover, there is no possibility that a substrate front face may be damaged, and since a substrate is not directly put to the plasma like the sputtering method, when this point produces a device to ***** on a semi-conductor substrate and it is necessary to make a dielectric thin film deposit, it is advantageous.

[0028] Furthermore, even if it is the case where it deposits again by making substrate temperature into temperature with low 200 [**] thru/or 450 [**] extent, it comes out practically, and sufficient electrical characteristics, i.e., a dielectric constant, are suitably high, a dielectric thin film with little leakage current can be obtained, and it is advantageous as compared with a sol-gel method.

[0029]

[Example] Drawing 1 is an important section explanatory view showing an example of the dielectric thin film deposition equipment used for carrying out this invention.

[0030] In drawing 1 Ion Gang and 3 for a raw material evaporation source and 2 Electronic irradiation equipment, In 4, a substrate maintenance base and 5 a heater and 8 for a substrate maintenance base slewing gear and 7 A vacuum evaporatio room, A shutter and 14 a crucible and 12 for the thickness gage for which 9 used the quartz resonator, and 11 An electron gun, In the power source for plasma generating, and 81, a plasma generating room and 82 show an introductory way, and 83 shows [the electrode for plasma generating with which a semi-conductor substrate and 61 make a gate valve, and, as for 71, 41 makes the shape of a ring, and 72] the electrode for plasma generating of an parallel monotonous mold, respectively.

[0031] It sets to the dielectric thin film deposition equipment of illustration, and is SrTiO₃ as a raw material to the raw material evaporation source 1. It puts in, and an electron ray is irradiated, carry out heating evaporation, the front face of the semi-conductor substrate 41 set to the substrate maintenance base 4 is made to vapor-deposit a dielectric thin film, and an oxygen molecule or an oxygen atom is irradiated at the semi-conductor substrate 41 in the case of the vacuum evaporatio (henceforth an oxygen exposure).

[0032] Since the oxide thin film obtained will usually be in a hypoxia condition when forming an oxide thin film in vacuum evaporatio therefore under low voltage, this oxygen exposure is carried out in order to compensate this.

[0033] An oxygen exposure is performed as follows. First, oxygen is introduced into Ion Gang 2, an oxygen atom or molecular ion is generated, this is accelerated in Ion Gang 2, and an oxygen atom or a molecular ion style is made to generate. Irradiate the electron ray which this ionic current was made to generate with electronic irradiation equipment 3, neutralize it electrically, an oxygen atom or a molecular flow (henceforth an oxygen style) is made to generate, and it is irradiated at the semi-conductor substrate 41.

[0034] The oxygen plasma besides the approach of irradiating the oxygen accelerated as mentioned above as a means with which oxygen is compensated can be used. For example, a RF etc. is fed from the power source 72 for plasma generating to the electrode 71 for plasma generating in the vacuum evaporatio room 8, and a dielectric can be vapor-deposited, generating the oxygen plasma in the

vacuum evaporation room 8.

[0035] Moreover, the oxygen plasma is generated at the plasma generating room 81 which is separate [the vacuum evaporation room 8], and a life feeds only long active oxygen into the vacuum evaporation room 8 through the introductory way 82, and may be made to vapor-deposit a dielectric.

[0036] In order to make the crystallinity of a dielectric thin film good in the case of vacuum evaporation, therefore, the semi-conductor substrate 41 is heated at a heater 7, and it is made suitable temperature.

[0037] above SrTiO₃ not only -- therefore, the dielectric which consists of mixed crystal, such as BaTiO₃, TiO₂, Ta₂O₅, PbTiO₃, PbZrO₃, or this, can also be vapor-deposited to the same approach as the above, and can be used as a thin film.

[0038] The deposition equipment of illustration is used for below and it is SrTiO₃. The case where a thin film is made to deposit is explained.

[0039] (1) Deposit a metal on the semi-conductor substrates 41, such as Si or GaAs, and form an electrode.

[0040] This electrode carries out the laminating of the thin film of Pt etc., and, specifically, is constituted. Pt used here has low resistance, it is excellent in thermal resistance, and adhesion with the semi-conductor substrate 41 or a high dielectric constant thin film, or a high dielectric film can substitute a good metal for it.

[0041] (2) SrTiO₃ which is a raw material in the crucible [in / it equips with the semi-conductor substrate 41 so that an electrode side may be expressed on the substrate maintenance base 4 in deposition equipment, and / the single raw material evaporation source 1] 11 It puts in.

[0042] (3) Operate an exhaustor 6, exhaust the inside of the vacuum evaporation room 8, and set a pressure to 1×10^{-5} [Torr] thru/or 1×10^{-2} [Torr]. Under the present circumstances, in the vacuum evaporation room 8, a pressure is adjusted by introducing into a mass flow controller the oxygen which therefore controlled the flow rate.

[0043] (4) A heater 7 is operated and let temperature of the semi-conductor substrate 41 be the range of 200 [°C] thru/or 450 [°C]. Under the present circumstances, in order to bring a temperature rise forward, you may heat auxiliary using a lamp.

[0044] (5) If the semi-conductor substrate 41 reaches predetermined temperature and the inside of the vacuum evaporation room 8 reaches a predetermined pressure, the electron beam from an electron gun 14 will be irradiated, and SrTiO₃ in a crucible 11 will be fused.

[0045] (6) In the case of vacuum evaporation, a shutter 12 is opened, start vacuum evaporation, operate the substrate maintenance base slewing gear 5, and rotate the substrate maintenance base 4.

[0046] SrTiO₃ deposited on the semi-conductor substrate 41 The monitor of the rate of sedimentation and thickness of a thin film is carried out by the thickness gage 9 which used the quartz resonator, and they feed this back to the control system of the electron gun 14 in the raw material evaporation source 1, and they control electron ray reinforcement automatically so that the rate of sedimentation becomes fixed.

[0047] Since the pressure in the vacuum evaporation room 8 is therefore controlled by installation of oxygen, the ambient atmosphere in the vacuum evaporation room 8 at the time of vacuum evaporation is mainly oxygen.

[0048] A neutrality oxygen style is electrically made from an operation of Ion Gang 2 and electronic irradiation equipment 3 to vacuum evaporation and coincidence, and this is irradiated at the semi-conductor substrate 41. It was made for the oxygen exposure in this case to become more than 1×10^{13} [/cm² and s] on the front face of the semi-conductor substrate 41.

[0049] When a dielectric thin film was formed in said process, the pressure and the rate of sedimentation in the vacuum evaporation room 8 were set constant, and the relation of the specific inductive capacity in an oxygen exposure and the dielectric thin film made to deposit was investigated.

[0050] Drawing 2 is a diagram showing the relation of the specific inductive capacity in the dielectric thin film made to deposit on an oxygen exposure list, and is an oxygen exposure in an axis of abscissa. On the axis of ordinate, the specific inductive capacity of a dielectric thin film is taken for 10^{13} [/cm²

and s] again, respectively.

[0051] The conditions at the time of obtaining the data looked at by drawing 2 are the cases where the pressure in the vacuum evaporation room 8 and the rate of sedimentation of a dielectric thin film are set constant.

[0052] It is ****(ed) that specific inductive capacity is so large that there are many exposures of oxygen so that clearly from drawing. This is considered because the presentation of the deposited dielectric thin film will be in the condition that oxygen is insufficient, as compared with a stoichiometry-presentation.

[0053] SrTiO₃ which the pressure in an oxygen exposure and the vacuum evaporation room 8 was set constant, and the rate of sedimentation was changed by changing the amount of electron beam irradiation of the raw material evaporation source 1, and was obtained when a dielectric thin film was formed in said process. The specific inductive capacity of a thin film was investigated.

[0054] Drawing 3 is a diagram showing the relation of the specific inductive capacity in the dielectric thin film made to deposit on a rate-of-sedimentation list, and the rate of sedimentation is taken on an axis of abscissa, and it has taken the specific inductive capacity of a dielectric thin film on the axis of ordinate, respectively. In addition, this data is obtained considering an oxygen exposure as fixed.

[0055] In drawing, when the oxygen tension of a white round head is about 1×10^{-3} [Torr], a black dot is the case where oxygen tension is about 1×10^{-4} [Torr].

[0056] When it becomes below 0.05 [nm/s] especially as the rate of sedimentation becomes smaller than 0.2 [nm] so that clearly from drawing, specific inductive capacity is increasing. It is thought that this cause is for the oxygen supply per unit time amount to decrease effectually since the oxygen of the deposited thin film is insufficient when the rate of sedimentation is slow, or is because the crystallinity of the thin film deposited when the rate of sedimentation was slow is good.

[0057] In the thin film which the rate of sedimentation was changed and was made to deposit, there was almost no presentation change of Sr, Ti, and O which depend on having changed the rate of sedimentation in the result of having investigated the presentation of Sr, Ti, and O by X-ray photoelectron spectroscopy (X-ray photoelectron spectroscopy: XPS) and electron ray probe microanalyzer (electron probe microanalyzer: EPMA).

[0058] Ti and Sr full width at half maximum in XPS were small in the thin film with the rate of sedimentation in a thin film with the large rate of sedimentation, it is large, and small. It is thought that the crystallinity of the deposited thin film improved and specific inductive capacity improved, so that the rate of sedimentation is small, since the crystallinity of the deposited thin film is generally so good that half-value width is small.

[0059] When a dielectric thin film was formed in said process, the relation of the specific inductive capacity in the pressure and the dielectric thin film made to deposit in the vacuum evaporation room 8 at the time of deposition was investigated.

[0060] Drawing 4 is a diagram showing the relation of the specific inductive capacity in the dielectric thin film made to deposit on the pressure list of the vacuum evaporation interior of a room, and has taken the specific inductive capacity of a dielectric thin film for the pressure of the vacuum evaporation interior of a room on the axis of abscissa again at the axis of ordinate, respectively. In addition, this data is obtained considering the rate of sedimentation and an oxygen exposure as fixed.

[0061] It is so large that a pressure is high, although there are few dependencies [on 1×10^{-5} [Torr] thru/or 1×10^{-2} [Torr] and as opposed to / pressure / a pressure in the specific inductive capacity of a dielectric thin film] so that clearly from drawing.

[0062] SrTiO₃ made to deposit from the above mentioned place. Specific inductive capacity is 40, leakage current is 10^{-8} [A/cm²] thru/or 10^{-7} [A/cm²] extent, and practical use can fully be presented with it.

[0063] At the above mentioned example, it is SrTiO₃. Although deposition of a thin film was explained, it is checking that the dielectric which consists of mixed crystal, such as BaTiO₃, TiO₂, Ta₂O₅, PbTiO₃, PbZrO₃, or this, can be similarly deposited in vacuum evaporation.

[0064] If it depends on the deposition equipment used in said example, deposition processing of a diameter 75 [mm] thru/or many semi-conductor substrates of 100 [mm] can be carried out collectively,

and there are very little thickness of the dielectric thin film in that case and variation of specific inductive capacity.

[0065] From the deposition technique of the interlayer insulation film which applied the usual ion beam assistant vacuum deposition, or the deposition technique of the thin film for optics, it was not just able to predict the above mentioned result at all.

[0066] Namely, it is common to make it deposit by setting the rate of sedimentation to 0.2 [nm/s] thru/or 0.5 [nm/s], and, moreover, it is known for thin films, such as this, that the dependency over the rate of sedimentation will hardly become a problem.

[0067] Moreover, in deposition of the interlayer insulation film which depends on ion beam assistant vacuum deposition, or the thin film for optics, since oxygen is irradiated directly at a substrate, a thin film [made / for oxygen tension to fall below to for example, about 10-4 [Torr] to some extent / better / crystallinity] can be obtained. That is, a good result is obtained for the direction which the accelerated oxygen made the oxygen tension of extent which does not collide with the oxygen in an ambient atmosphere.

[0068] However, in this invention, it will be understood that the result of having completely differed from the result obtained with the deposition technique of said interlayer insulation film or the deposition technique of the thin film for optics is obtained.

[0069] In this invention, without being restricted to said example, otherwise many alterations are possible, next it is illustrated.

[0070] It sets in said each example and is SrTiO₃. Although the case where the accelerated oxygen molecule was used was explained when making it deposit, it replaces with this, and it can also vapor-deposit, generating the oxygen plasma in the vacuum evaporation room 8. In that case, the oxygen tension and evaporation rate in the vacuum evaporation room 8 may be chosen as the same range as said each example.

[0071] From the power source 72 for plasma generating, generating of the oxygen plasma sets 300 [W] thru/or 500 [W], and a frequency to 13.56 [GHz] for power, feeds a RF into the electrode 71 for plasma generating in the vacuum evaporation room 8, and generates the oxygen plasma.

[0072] Deposited SrTiO₃ Like said example, the electrical characteristics of a thin film were good, when the rate of sedimentation was below 0.2 [nm/s], or when oxygen tension was 10-4 [Torr] thru/or 10-3 [Torr].

[0073] Moreover, it is SrTiO₃ like said each example, generating the oxygen plasma at the oxygen plasma generating room 81 which is the vacuum evaporation room 8 and another room, and feeding active oxygen into the vacuum evaporation room 8 through the introductory way 82. You may vapor-deposit.

[0074] From the power source 72 for plasma generating, generating of the oxygen plasma sets 300 [W] thru/or 500 [W], and a frequency to 13.56 [GHz] for power, feeds a RF into the electrode 73 for plasma generating of the parallel monotonous mold in the oxygen plasma generating room 81, and generates the oxygen plasma.

[0075] In this case, SrTiO₃ deposited even if set Like said example, the electrical characteristics of a thin film were good, when the rate of sedimentation was below 0.2 [nm/s], or when oxygen tension was 10-4 [Torr] thru/or 10-3 [Torr].

[0076]

[Effect of the Invention] An oxide quantity dielectric constant thin film or an oxide ferroelectric thin film is made to vapor-deposit and deposit on a substrate as below the rate of sedimentation 0.2 [nm/s] by the dielectric thin film deposition approach which depends on this invention under the ambient atmosphere containing active oxygen, such as an accelerated oxygen molecule, an oxygen radical, an oxygen atom, or oxygen plasma.

[0077] A thing it is [leakage current] able to form easily few high dielectric constant thin films or a ferroelectric thin film in taking said configuration highly [specific inductive capacity] therefore as compared with the usual vacuum deposition it, and good to extent which is equal to the use as a capacitor for integrated circuits enough can be obtained.

[0078] Moreover, since vacuum deposition is applicable, many diameter semi-conductor substrates of macrostomia can be processed collectively and homogeneous deposition can moreover be performed in a short time, as compared with the sputtering method, it excels remarkably in respect of mass-production nature.

[0079] Moreover, a thin film can be made to deposit, without degrading the device property from the ability processing at low temperature as compared with a sol-gel method, also after producing a transistor, for example to a semi-conductor substrate.

[0080] moreover, like [in the case of depending on a CVD method], Sr and the raw material of Ba are difficult to receive, and expensive -- etc. -- it is satisfactory.

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TECHNICAL FIELD

[Industrial Application] This invention relates to the approach of depositing a suitable dielectric thin film producing a small capacitor for example, to a semi-conductor substrate etc.

[0002] It will be necessary to make small the capacitor of a comparatively big capacity connected out of the chip, and it will need to be incorporated as an internal component of MMIC in the microwave integrated circuit (monolithic microwave integrated circuit:MMIC) for recent years, for example, cellular phones, etc. for a miniaturization or low-pricing.

[0003] Moreover, like dynamic random access memory (dynamic random access memory:DRAM), detailed-ization is needed with high integration, and it is necessary to produce the capacitor for storage maintenance in a small area.

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PRIOR ART

[Description of the Prior Art] Usually, the high dielectric constant for capacitors The ingredient which it has or a ferroelectric 3, for example, SrTiO₃, BaTiO₃, TiO₂, Ta₂O₅, PbTiO₃, and PbZrO₃ Or mixed crystal, such as this, etc. For example, the sputtering method (see "JP,58-194206,A", if it requires), a chemical-vapor-deposition method (if it requires) H. "Yamaguchi et al., Jpn.J.Appl.Phys.vol.32 No.9B, pp.4069-4073 (1993)" -- reference and a sol-gel method (if it requires) Therefore, it is produced by applying [reference] "JP,56-28408,A."

[0006] By the sputtering method in said Prior art, the dielectric thin film is formed by making it for example, low-pressure Ar ambient atmosphere, generating the plasma and generally, carrying out sputtering of the target within a vacuum housing, after setting the semi-conductor substrate which should counter the target which consists of a dielectric raw material, and should form a dielectric thin film.

[0007] In this case, there is comparatively little number of sheets of the semi-conductor substrate which can be processed at once since there is a limitation in the magnitude of a target, and it has a possibility of deteriorating the property of the device which a semi-conductor substrate front face is put to the ion of high energy etc., and is easy to damage, therefore is made in order to form a thin film in the plasma.

[0008] moreover, chemical vapor deposition (chemical vapor deposition:CVD) -- although material gas is made to react at an elevated temperature and a dielectric thin film is made to deposit on a semi-conductor substrate in law -- SrTiO₃ and BaTiO₃ etc. -- forming thin films, such as this, on mass-production level, since there are not Sr raw material with high vapor pressure and a Ba raw material in order to make it grow up -- in practice -- coming out -- it is difficult.

[0009] Furthermore, apply the emulsion or suspension which consists of an organic solvent and dielectric-materials impalpable powder on a semi-conductor substrate, and therefore evaporate an organic solvent to process this at the elevated temperature more than about 600 [**], and he makes a particle sinter, and is trying to obtain a desired dielectric thin film with a sol-gel method again.

[0010] Therefore, in the phase after forming a transistor, although the processing in an elevated temperature is indispensable, when an elevated-temperature process cannot be applied, for example, since a transistor deteriorates by applying an elevated temperature, this means cannot be adopted.

[0011] By the way, a lot of [faults / there are comparatively few faults looked at by the above mentioned all directions method, namely,] semi-conductor substrates can be made to deposit an oxide thin film at low temperature, and vacuum deposition is known as an approach with doing [little] damage to a semi-conductor substrate front face like the sputtering method.

[0012] However, the oxygen in the oxide thin film made to deposit when making an oxide thin film deposit, in order to carry out in a vacuum tends to escape from vacuum deposition, and a good thin film is not obtained.

[0013] The approach of vapor-depositing like for example, ion beam assistant vacuum deposition, there, while irradiating an oxygen beam at a thin film is also proposed.

[0014] However, it is [that the thin film for optics (see "JP,61-288064,A" if it requires), and the interlayer insulation film (see "JP,3-196526,A" if it requires) in an integrated circuit are generally only

known for the thin film for capacitors as an oxide thin film formed with the application of ion beam assistant vacuum deposition since it is required that electrical characteristics' being good, especially specific inductive capacity should be high, and leakage current should be low, and], and there is no example about the thin film for capacitors.

[0015] Therefore, this invention persons are actually SrTiO_3 , BaTiO_3 , TiO_2 , Ta_2O_5 , PbTiO_3 , and PbZrO_3 to the vacuum deposition indicated by said each patent official report. Although the capacitor was made as an experiment using the thin film, no thin film was able to fulfill high specific inductive capacity and the conditions of little leakage current.

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EFFECT OF THE INVENTION

[Effect of the Invention] An oxide quantity dielectric constant thin film or an oxide ferroelectric thin film is made to vapor-deposit and deposit on a substrate as below the rate of sedimentation 0.2 [nm/s] by the dielectric thin film deposition approach which depends on this invention under the ambient atmosphere containing active oxygen, such as an accelerated oxygen molecule, an oxygen radical, an oxygen atom, or oxygen plasma.

[0077] A thing it is [leakage current] able to form easily few high dielectric constant thin films or a ferroelectric thin film in taking said configuration highly [specific inductive capacity] therefore as compared with the usual vacuum deposition it, and good to extent which is equal to the use as a capacitor for integrated circuits enough can be obtained.

[0078] Moreover, since vacuum deposition is applicable, many diameter semi-conductor substrates of macrostomia can be processed collectively and homogeneous deposition can moreover be performed in a short time, as compared with the sputtering method, it excels remarkably in respect of mass-production nature.

[0079] Moreover, a thin film can be made to deposit, without degrading the device property from the ability processing at low temperature as compared with a sol-gel method, also after producing a transistor, for example to a semi-conductor substrate.

[0080] moreover, like [in the case of depending on a CVD method], Sr and the raw material of Ba are difficult to receive, and expensive -- etc. -- it is satisfactory.

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TECHNICAL PROBLEM

[Problem(s) to be Solved by the Invention] It enables it to mass-produce a thin film with the high dielectric constant which can be used for the capacitor for integrated circuits at comparatively low substrate temperature by this invention. For this reason, vacuum deposition is applied, electrical characteristics were excellent, namely, specific inductive capacity makes high deposition of a high dielectric thin film with little leakage current possible.

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MEANS

[Means for Solving the Problem] This invention is selecting the rate of sedimentation and the oxygen tension at the time of deposition to a suitable value, when making a dielectric thin film deposit with the application of vacuum deposition with the compensation means of oxygen like ion beam assistant vacuum deposition, and it has been to the base to realize a dielectric thin film with the outstanding property which is equal to use as a configuration member of the capacitor for integrated circuits.

[0018] In the dielectric thin film deposition approach which depends on this invention from the above mentioned place (1) The accelerated oxygen molecule, an oxygen radical, an oxygen atom, Or [whether it is characterized by making an oxide quantity dielectric constant thin film or an oxide ferroelectric thin film (for example, SrTiO₃ thin film etc.) vapor-deposit and deposit on a substrate (for example, semiconductor substrate 41) as below the rate of sedimentation 0.2 [nm/s] under the ambient atmosphere containing active oxygen (oxygen style), such as oxygen plasma, and] or [0019] (2) or [that it is characterized by carrying out the oxygen tension in an ambient atmosphere more than 10⁻⁴ [Torr] in the above (1)] -- or [0020] (3) the above (1) or (2) -- setting -- an oxide quantity dielectric constant thin film or an oxide ferroelectric thin film -- SrTiO₃, BaTiO₃, TiO₂, Ta₂O₅, PbTiO₃, and PbZrO₃ any -- or [or / that it is characterized by being mixed crystal, such as this,] -- or [0021] (4) The oxygen molecule accelerated in the above (1), (2), or (3), An oxygen radical, or [that it is characterized by for an electron (for example, electron from electronic irradiation equipment 3) neutralizing electrically the oxygen ion with which the oxygen atom etc. was accelerated by Ion Gang (for example, Ion Gang 2), and being generated, and irradiating more than 1x10¹³ [/cm² and s] to a substrate] -- or [0022] (5) or [that it is characterized by generating the oxygen plasma at a vacuum evaporatio room in the above (1), (2), or (3)] -- or [0023] (6) or [that it is characterized by introducing into a vacuum evaporatio room the active oxygen which is made to generate the oxygen plasma and is obtained in the above (1), (2), or (3) at an oxygen plasma generating room] -- or [0024] (7) In the above (1), (2), (3), (4), (5), or (6), substrate temperature is carried out more than 200 [**], and it is characterized by making an oxide quantity dielectric constant thin film or an oxide ferroelectric thin film vapor-deposit and deposit.

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OPERATION

[Function] A diameter is able to be able to use a thing large-sized as deposition equipment for it, and to carry out batch processing of many semi-conductor substrates of 75 [mm] thru/or 100 [mm] to it therefore, since it is the same as carrying out the usual vacuum evaporationo for taking said means fundamentally.

[0026] Moreover, as for a different place from the usual vacuum evaporationo, since the amount of oxygen therefore contained in an oxygen exposure at a dielectric thin film is controllable, electrical characteristics can make a good dielectric thin film able to deposit, and can raise the crystallinity of a dielectric thin film by therefore making a substrate a heater at suitable temperature in the case of vacuum evaporationo.

[0027] Moreover, there is no possibility that a substrate front face may be damaged, and since a substrate is not directly put to the plasma like the sputtering method, when this point produces a device to ***** on a semi-conductor substrate and it is necessary to make a dielectric thin film deposit, it is advantageous.

[0028] Furthermore, even if it is the case where it deposits again by making substrate temperature into temperature with low 200 [**] thru/or 450 [**] extent, it comes out practically, and sufficient electrical characteristics, i.e., a dielectric constant, are suitably high, a dielectric thin film with little leakage current can be obtained, and it is advantageous as compared with a sol-gel method.

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EXAMPLE

[Example] Drawing 1 is an important section explanatory view showing an example of the dielectric thin film deposition equipment used for carrying out this invention.

[0030] In drawing 1 Ion Gang and 3 for a raw material evaporation source and 2 Electronic irradiation equipment, In 4, a substrate maintenance base and 5 a heater and 8 for a substrate maintenance base slewing gear and 7 A vacuum evaporatio room, A shutter and 14 a crucible and 12 for the thickness gage for which 9 used the quartz resonator, and 11 An electron gun, In the power source for plasma generating, and 81, a plasma generating room and 82 show an introductory way, and 83 shows [the electrode for plasma generating with which a semi-conductor substrate and 61 make a gate valve, and, as for 71, 41 makes the shape of a ring, and 72] the electrode for plasma generating of an parallel monotonous mold, respectively.

[0031] It sets to the dielectric thin film deposition equipment of illustration, and is SrTiO₃ as a raw material to the raw material evaporation source 1. It puts in, and an electron ray is irradiated, carry out heating evaporation, the front face of the semi-conductor substrate 41 set to the substrate maintenance base 4 is made to vapor-deposit a dielectric thin film, and an oxygen molecule or an oxygen atom is irradiated at the semi-conductor substrate 41 in the case of the vacuum evaporatio (henceforth an oxygen exposure).

[0032] Since the oxide thin film obtained will usually be in a hypoxia condition when forming an oxide thin film in vacuum evaporatio therefore under low voltage, this oxygen exposure is carried out in order to compensate this.

[0033] An oxygen exposure is performed as follows. First, oxygen is introduced into Ion Gang 2, an oxygen atom or molecular ion is generated, this is accelerated in Ion Gang 2, and an oxygen atom or a molecular ion style is made to generate. Irradiate the electron ray which this ionic current was made to generate with electronic irradiation equipment 3, neutralize it electrically, an oxygen atom or a molecular flow (henceforth an oxygen style) is made to generate, and it is irradiated at the semi-conductor substrate 41.

[0034] The oxygen plasma besides the approach of irradiating the oxygen accelerated as mentioned above as a means with which oxygen is compensated can be used. For example, a RF etc. is fed from the power source 72 for plasma generating to the electrode 71 for plasma generating in the vacuum evaporatio room 8, and a dielectric can be vapor-deposited, generating the oxygen plasma in the vacuum evaporatio room 8.

[0035] Moreover, the oxygen plasma is generated at the plasma generating room 81 which is separate [the vacuum evaporatio room 8], and a life feeds only long active oxygen into the vacuum evaporatio room 8 through the introductory way 82, and may be made to vapor-deposit a dielectric.

[0036] In order to make the crystallinity of a dielectric thin film good in the case of vacuum evaporatio, therefore, the semi-conductor substrate 41 is heated at a heater 7, and it is made suitable temperature.

[0037] above SrTiO₃ not only -- therefore, the dielectric which consists of mixed crystal, such as BaTiO₃, TiO₂, Ta₂O₅, PbTiO₃, PbZrO₃, or this, can also be vapor-deposited to the same approach as

the above, and can be used as a thin film.

[0038] The deposition equipment of illustration is used for below and it is SrTiO₃. The case where a thin film is made to deposit is explained.

[0039] (1) Deposit a metal on the semi-conductor substrates 41, such as Si or GaAs, and form an electrode.

[0040] This electrode carries out the laminating of the thin film of Pt etc., and, specifically, is constituted. Pt used here has low resistance, it is excellent in thermal resistance, and adhesion with the semi-conductor substrate 41 or a high dielectric constant thin film, or a high dielectric film can substitute a good metal for it.

[0041] (2) SrTiO₃ which is a raw material in the crucible [in / it equips with the semi-conductor substrate 41 so that an electrode side may be expressed on the substrate maintenance base 4 in deposition equipment, and / the single raw material evaporation source 1] 11 It puts in.

[0042] (3) Operate an exhauster 6, exhaust the inside of the vacuum evaporatio room 8, and set a pressure to 1×10^{-5} [Torr] thru/or 1×10^{-2} [Torr]. Under the present circumstances, in the vacuum evaporatio room 8, a pressure is adjusted by introducing into a mass flow controller the oxygen which therefore controlled the flow rate.

[0043] (4) A heater 7 is operated and let temperature of the semi-conductor substrate 41 be the range of 200 [°C] thru/or 450 [°C]. Under the present circumstances, in order to bring a temperature rise forward, you may heat auxiliary using a lamp.

[0044] (5) If the semi-conductor substrate 41 reaches predetermined temperature and the inside of the vacuum evaporatio room 8 reaches a predetermined pressure, the electron beam from an electron gun 14 will be irradiated, and SrTiO₃ in a crucible 11 will be fused.

[0045] (6) In the case of vacuum evaporatio, a shutter 12 is opened, start vacuum evaporatio, operate the substrate maintenance base slewing gear 5, and rotate the substrate maintenance base 4.

[0046] SrTiO₃ deposited on the semi-conductor substrate 41 The monitor of the rate of sedimentation and thickness of a thin film is carried out by the thickness gage 9 which used the quartz resonator, and they feed this back to the control system of the electron gun 14 in the raw material evaporation source 1, and they control electron ray reinforcement automatically so that the rate of sedimentation becomes fixed.

[0047] Since the pressure in the vacuum evaporatio room 8 is therefore controlled by installation of oxygen, the ambient atmosphere in the vacuum evaporatio room 8 at the time of vacuum evaporatio is mainly oxygen.

[0048] A neutrality oxygen style is electrically made from an operation of Ion Gang 2 and electronic irradiation equipment 3 to vacuum evaporatio and coincidence, and this is irradiated at the semi-conductor substrate 41. It was made for the oxygen exposure in this case to become more than 1×10^{13} [/cm² and s] on the front face of the semi-conductor substrate 41.

[0049] When a dielectric thin film was formed in said process, the pressure and the rate of sedimentation in the vacuum evaporatio room 8 were set constant, and the relation of the specific inductive capacity in an oxygen exposure and the dielectric thin film made to deposit was investigated.

[0050] Drawing 2 is a diagram showing the relation of the specific inductive capacity in the dielectric thin film made to deposit on an oxygen exposure list, and is an oxygen exposure in an axis of abscissa. On the axis of ordinate, the specific inductive capacity of a dielectric thin film is taken for 10^{13} [/cm² and s] again, respectively.

[0051] The conditions at the time of obtaining the data looked at by drawing 2 are the cases where the pressure in the vacuum evaporatio room 8 and the rate of sedimentation of a dielectric thin film are set constant.

[0052] It is ****(ed) that specific inductive capacity is so large that there are many exposures of oxygen so that clearly from drawing. This is considered because the presentation of the deposited dielectric thin film will be in the condition that oxygen is insufficient, as compared with a stoichiometry-presentation.

[0053] SrTiO₃ which the pressure in an oxygen exposure and the vacuum evaporatio room 8 was set constant, and the rate of sedimentation was changed by changing the amount of electron beam

irradiation of the raw material evaporation source 1, and was obtained when a dielectric thin film was formed in said process. The specific inductive capacity of a thin film was investigated.

[0054] Drawing 3 is a diagram showing the relation of the specific inductive capacity in the dielectric thin film made to deposit on a rate-of-sedimentation list, and the rate of sedimentation is taken on an axis of abscissa, and it has taken the specific inductive capacity of a dielectric thin film on the axis of ordinate, respectively. In addition, this data is obtained considering an oxygen exposure as fixed.

[0055] In drawing, when the oxygen tension of a white round head is about 1×10^{-3} [Torr], a black dot is the case where oxygen tension is about 1×10^{-4} [Torr].

[0056] When it becomes below 0.05 [nm/s] especially as the rate of sedimentation becomes smaller than 0.2 [nm/s] so that clearly from drawing, specific inductive capacity is increasing. It is thought that this cause is for the oxygen supply per unit time amount to decrease effectually since the oxygen of the deposited thin film is insufficient when the rate of sedimentation is slow, or is because the crystallinity of the thin film deposited when the rate of sedimentation was slow is good.

[0057] In the thin film which the rate of sedimentation was changed and was made to deposit, there was almost no presentation change of Sr, Ti, and O which depend on having changed the rate of sedimentation in the result of having investigated the presentation of Sr, Ti, and O by X-ray photoelectron spectroscopy (X-ray photoelectron spectroscopy: XPS) and electron ray probe microanalyzer (electron probe microanalyzer: EPMA).

[0058] Ti and Sr full width at half maximum in XPS were small in the thin film with the rate of sedimentation in a thin film with the large rate of sedimentation, it is large, and small. It is thought that the crystallinity of the deposited thin film improved and specific inductive capacity improved, so that the rate of sedimentation is small, since the crystallinity of the deposited thin film is generally so good that half-value width is small.

[0059] When a dielectric thin film was formed in said process, the relation of the specific inductive capacity in the pressure and the dielectric thin film made to deposit in the vacuum evaporation room 8 at the time of deposition was investigated.

[0060] Drawing 4 is a diagram showing the relation of the specific inductive capacity in the dielectric thin film made to deposit on the pressure list of the vacuum evaporation interior of a room, and has taken the specific inductive capacity of a dielectric thin film for the pressure of the vacuum evaporation interior of a room on the axis of abscissa again at the axis of ordinate, respectively. In addition, this data is obtained considering the rate of sedimentation and an oxygen exposure as fixed.

[0061] It is so large that a pressure is high, although there are few dependencies [on 1×10^{-5} [Torr] thru/or 1×10^{-2} [Torr] and as opposed to / pressure / a pressure in the specific inductive capacity of a dielectric thin film] so that clearly from drawing.

[0062] SrTiO₃ made to deposit from the above mentioned place. Specific inductive capacity is 40, leakage current is 10^{-8} [A/cm²] thru/or 10^{-7} [A/cm²] extent, and practical use can fully be presented with it.

[0063] At the above mentioned example, it is SrTiO₃. Although deposition of a thin film was explained, it is checking that the dielectric which consists of mixed crystal, such as BaTiO₃, TiO₂, Ta₂O₅, PbTiO₃, PbZrO₃, or this, can be similarly deposited in vacuum evaporation.

[0064] If it depends on the deposition equipment used in said example, deposition processing of a diameter 75 [mm] thru/or many semi-conductor substrates of 100 [mm] can be carried out collectively, and there are very little thickness of the dielectric thin film in that case and variation of specific inductive capacity.

[0065] From the deposition technique of the interlayer insulation film which applied the usual ion beam assistant vacuum deposition, or the deposition technique of the thin film for optics, it was not just able to predict the above mentioned result at all.

[0066] Namely, it is common to make it deposit by setting the rate of sedimentation to 0.2 [nm/s] thru/or 0.5 [nm/s], and, moreover, it is known for thin films, such as this, that the dependency over the rate of sedimentation will hardly become a problem.

[0067] Moreover, in deposition of the interlayer insulation film which depends on ion beam assistant

vacuum deposition, or the thin film for optics, since oxygen is irradiated directly at a substrate, a thin film [made / for oxygen tension to fall below to for example, about 10^{-4} [Torr] to some extent / better / crystallinity] can be obtained. That is, a good result is obtained for the direction which the accelerated oxygen made the oxygen tension of extent which does not collide with the oxygen in an ambient atmosphere.

[0068] However, in this invention, it will be understood that the result of having completely differed from the result obtained with the deposition technique of said interlayer insulation film or the deposition technique of the thin film for optics is obtained.

[0069] In this invention, without being restricted to said example, otherwise many alterations are possible, next it is illustrated.

[0070] It sets in said each example and is SrTiO_3 . Although the case where the accelerated oxygen molecule was used was explained when making it deposit, it replaces with this, and it can also vapor-deposit, generating the oxygen plasma in the vacuum evaporation room 8. In that case, the oxygen tension and evaporation rate in the vacuum evaporation room 8 may be chosen as the same range as said each example.

[0071] From the power source 72 for plasma generating, generating of the oxygen plasma sets 300 [W] thru/or 500 [W], and a frequency to 13.56 [GHz] for power, feeds a RF into the electrode 71 for plasma generating in the vacuum evaporation room 8, and generates the oxygen plasma.

[0072] Deposited SrTiO_3 Like said example, the electrical characteristics of a thin film were good, when the rate of sedimentation was below 0.2 [nm/s], or when oxygen tension was 10^{-4} [Torr] thru/or 10^{-3} [Torr].

[0073] Moreover, it is SrTiO_3 like said each example, generating the oxygen plasma at the oxygen plasma generating room 81 which is the vacuum evaporation room 8 and another room, and feeding active oxygen into the vacuum evaporation room 8 through the introductory way 82. You may vapor-deposit.

[0074] From the power source 72 for plasma generating, generating of the oxygen plasma sets 300 [W] thru/or 500 [W], and a frequency to 13.56 [GHz] for power, feeds a RF into the electrode 73 for plasma generating of the parallel monotonous mold in the oxygen plasma generating room 81, and generates the oxygen plasma.

[0075] In this case, SrTiO_3 deposited even if set Like said example, the electrical characteristics of a thin film were good, when the rate of sedimentation was below 0.2 [nm/s], or when oxygen tension was 10^{-4} [Torr] thru/or 10^{-3} [Torr].

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DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

[Drawing 1] It is an important section explanatory view showing an example of the dielectric thin film deposition equipment used for carrying out this invention.

[Drawing 2] It is a diagram showing the relation of the specific inductive capacity in the dielectric thin film made to deposit on an oxygen exposure list.

[Drawing 3] It is a diagram showing the relation of the specific inductive capacity in the dielectric thin film made to deposit on a rate-of-sedimentation list.

[Drawing 4] It is a diagram showing the relation of the specific inductive capacity in the dielectric thin film made to deposit on the pressure list of the vacuum evaporatio interior of a room.

[Description of Notations]

- 1 Raw Material Evaporation Source
- 2 Ion Gang
- 3 Electronic Irradiation Equipment
- 4 Substrate Maintenance Base
- 5 Substrate Maintenance Base Slewing Gear
- 7 Heater
- 8 Vacuum Evaporatio Room
- 9 Thickness Gage
- 11 Crucible
- 12 Shutter
- 14 Electron Gun
- 41 Semi-conductor Substrate
- 61 Gate Valve
- 71 Electrode for Plasma Generating
- 72 Power Source for Plasma Generating
- 81 Plasma Generating Room
- 82 Introductory Way
- 83 Electrode for Plasma Generating

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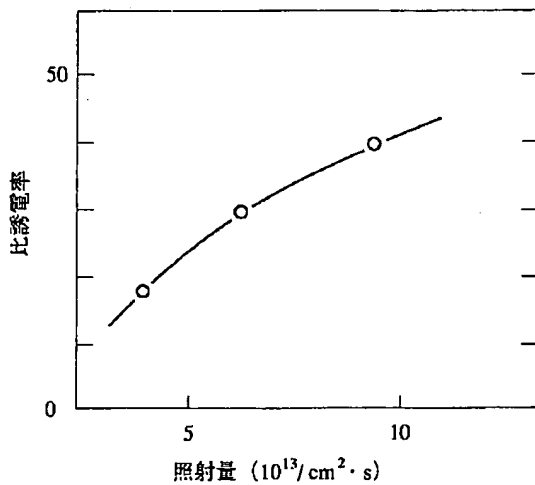
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DRAWINGS

[Drawing 2]

酸素照射量と比誘電率との関係を表す線図



[Drawing 3]

堆積速度と比誘電率との関係を表す線図

